Magic-wave-induced $^1S_0-^3P_0$ transition in even isotopes of alkaline-earth-like atoms

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The circular polarized laser beam of the “magic” wavelength may be used for mixing the $^3P_1$ state into the long-living metastable state $^3P_0$, thus enabling the strictly forbidden $^1S_0-^3P_0$ "clock" transition in even isotopes of alkaline-earth-like atoms, without change of the transition frequency. In odd isotopes the laser beam may adjust to an optimum value the line width of the "clock" transition, originally enabled by the hyperfine mixing. We present a detailed analysis of various factors influencing resolution and uncertainty for an optical frequency standard based on atoms exposed simultaneously to the lattice standing wave and an additional "state-mixing" wave, including estimations of the “magic” wavelengths, Rabi frequencies for the "clock" and state-mixing transitions, ac Stark shifts for the ground and metastable states of divalent atoms.

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Extremely narrow atomic line corresponding to a strictly forbidden $^1S_0-^3P_0$ transition between ground and metastable states of alkaline-earth-like atoms (such as Mg, Ca, Sr, Yb, Zn, Cd), currently considered as worthwhile candidates for an optical frequency standard, may be observed either on free odd isotopes [1, 2, 3] or on even isotopes in external fields [4, 5, 6, 7]. The mixing of the $^3P_1$ and $^3P_0$ states by the hyperfine interaction in the odd isotopes and by an external field in the even isotopes is the basic effect which removes the general selection-rule restrictions on the $0-0$ radiation transition. Intensive investigations of even alkaline-earth-like isotopes during the last few years were stimulated by a possibility to design a new frequency standard based on an oscillator with the record high quality Q-factor. In all the methods based on interrogation of the strongly forbidden $^1S_0-^3P_0$ transition in the even isotopes embedded into an optical lattice, engineered so as to equalize the upper- and lower-level Stark shifts, some additional radiation [4, 5] or static [6, 7] fields were applied.

In this article, we propose to use a circularly (elliptically) polarized wave of the “magic” wavelength λ_{mag} (corresponding to the so-called “Stark-cancellation” regime, see e.g. [1, 8]), in addition to the optical lattice field, in order to mix the $^3P_1$ state to the $^3P_0$ state. Since in even isotopes the nuclear momentum equals zero, both the initial and the final states of the frequency standard transition (the “clock” transition) have zero total momenta, without hyperfine structure splitting and without antisymmetric and tensor increments to the ac dipole polarizabilities and to the Stark effect. This makes the Stark shift of the upper and lower levels independent of polarization of external fields. Meanwhile the circular polarization of a laser wave allows for the second-order dipole-dipole mixing of the $^3P_1$ state to the metastable $^3P_0$ state, which is strictly forbidden for the linear polarization.

So, the role of the optical lattice field consists in trapping neutral atoms effectively free from collisions and Doppler effect (Lamb-Dicke regime) as well as from the light field perturbations [1], whereas an additional beam of the magic frequency, but with compulsory circular (elliptic) polarization, will enable the strictly forbidden radiation transitions via mixing the $^3P_1$ state to the metastable $^3P_0$ state. The two waves may be generated by one and the same laser or be completely independent, each properly adjusted to some particular conditions. So they may have different intensities, polarizations, wave vectors and even different wavelengths, subject, however, to the Stark-cancellation regime. In contrast with other methods [4]–[7], in this approach the atoms are exposed to only the magic-wavelength radiation, no additional ac or dc field is used and therefore no additional shift of the clock frequency can arise.

The origin of the laser radiation-induced mixing consists in the possibility of the second-order dipole transi-

tion between the $^3P_1$ state and the metastable $^3P_0$ state in ac field of a "magic" frequency $\omega{=}\omega_m{=}2\pi c/\lambda_{mag}$ (c is the speed of light) with a circular (elliptical) polarization. To this end, together with the standing wave of the optical lattice, a circularly polarized wave of the magic frequency should be used, which we further consider as the running wave with the electric field vector

$$\boldsymbol{F}_r(\boldsymbol{r},t) = F_r \mathrm{Re}\left\{\boldsymbol{e}\cdot\exp[i(\boldsymbol{k}\cdot\boldsymbol{r}-\omega_m t)]\right\}, \qquad (1)$$

where F_r is a real scalar amplitude, $\boldsymbol{e}$ is a complex unit polarization vector, $\boldsymbol{k} = \boldsymbol{n}\omega_m/c$ is the wave vector with the unit vector $\boldsymbol{n}$ which should have a non-zero component at right angle to the optical lattice beam in order that interrogation wave could travel along the lattice in compliance with the Doppler-cancellation conditions (for simplicity, we assume a 1D lattice here). The contribution of the $^3P_1$-state wave function into the metastable $^3P_0$-state wave function is determined by the ratio of the field-induced $^3P_0{-}^3P_1$ transition amplitude (Rabi frequency) W_{10} to the fine-structure splitting $\Delta_{10}{=}E_{^3P_1}{-}E_{^3P_0}$. In the nonrelativistic dipole approximation, the lowest non-vanishing (second) order in F_r amplitude (the atomic units are used in this paper, if not otherwise indicated)

$$W_{10} = -\frac{F_r^2}{4\sqrt{6}}\,\xi\,\alpha^a_{^3P}(\omega_m) \qquad (2)$$

is directly proportional to the circular polarization degree $\xi = i(\boldsymbol{n}\cdot[\boldsymbol{e}\times\boldsymbol{e}^*])$ and to the antisymmetric part $\alpha^a_{^3P}(\omega_m)$ of the ^3P_J triplet state ac polarizability, which e.g. for the state with maximal total momentum $J = L + S = 2$ is (see [9, 10, 11]):

$$\begin{aligned}\alpha_{^3PJM}(\omega) \;&=\; \alpha^s_{^3P}(\omega) + \frac{M}{2J}\xi\alpha^a_{^3P}(\omega)\\ &\quad -\frac{3M^2 - J(J+1)}{2J(2J-1)}\,\alpha^t_{^3P}(\omega),\end{aligned} \qquad (3)$$

here $M{=}(\boldsymbol{n}\cdot\boldsymbol{J})$ is the magnetic quantum number; the superscripts (s) and (t) indicate the scalar and tensor parts of the ac polarizability $\alpha_{^3PJM}(\omega)$.

Actually, the amplitude (2) may be compared to the amplitude of the hyperfine interaction, which mixes the states in the odd isotopes [8, 10], or to the magnetic-field-induced amplitude when the atoms experience the action of a magnetic field, which may also be used for the $^3P_0{-}^3P_1$ state mixing [6, 7]. Numerical computations carried out in the single-electron approximation with the use of the model potential method for analytical presentation of the radial wave functions [9, 10], gave the numerical values of the antisymmetric polarizabilities presented in Table I for Mg, Ca, Sr, Yb, Zn and Cd atoms at the "magic" wavelength corresponding to equal second-order ac Stark shifts $\Delta E(\,^3P_0) = \Delta E(\,^1S_0) = E^{(2)}_L$ of the metastable and ground states (the lattice depth)

$$E^{(2)}_L = -\frac{1}{4}\alpha^s(\omega_m)F_L^2, \qquad (4)$$

where $\alpha^s(\omega_m) = \alpha_{^1S_0}(\omega_m) = \alpha_{^3P_0}(\omega_m)$ is the ac polarizability of the "clock" levels; F_L represents the amplitude near the antinode of the lattice standing wave, oscillating with the "magic" frequency ω_m.

The Rabi frequency for the running-wave-induced transition (2) is directly proportional to the product of the wave intensity $I_r{=}cF_r^2/8\pi$ and to the antisymmetric polarizability $\alpha^a_{^3P}$, and may be presented in *MHz*, as follows

$$W_{10} = -0.01915\,\xi\alpha^a_{^3P}(\omega_m)I_r, \qquad (5)$$

where I_r is taken in MW/cm^2 and $\alpha^a_{^3P}$ in atomic units. The value of W_{10} determines the magnitude of the coefficient

$$a_1 = \frac{W_{10}}{\Delta_{10}} \qquad (6)$$

for the running-wave-induced contribution of the $^3P_1$ state to the wave function of an atom initially (when the field (1) is off) in the metastable $^3P_0$ state

$$\begin{aligned}|\psi\rangle \;&=\; |^3P_0\rangle + a_1|^3P_1\rangle\\ &= |^3P_0\rangle + a_1\left(a|^3P_1^{(0)}\rangle + b|^1P_1^{(0)}\rangle\right),\end{aligned} \qquad (7)$$

where the superscript (0) indicates a pure LS-state. The singlet-triplet mixing coefficients a and b in (7) may be calculated using the ratio of the lifetimes $\tau(\,^1P_1)$, $\tau(\,^3P_1)$ of singlet and triplet levels and the wavelengths $\lambda(\,^1P_1 - {}^1S_0)$, $\lambda(\,^3P_1 - {}^1S_0)$ of photons emitted in their radiation decay, as follows:

$$\frac{b^2}{a^2} = \frac{\tau(\,^1P_1)\lambda^3(\,^3P_1 - \,^1S_0)}{\tau(\,^3P_1)\lambda^3(\,^1P_1 - \,^1S_0)}, \quad a^2 + b^2 = 1. \qquad (8)$$

For I_r in MW/cm^2, Δ_{10} in cm^{-1} and $\alpha^a_{^3P}$ in atomic units the rate of the laser field-induced radiation transition $^3P_0{\to}^1S_0$ may be written as

$$w = |a_1|^2 w_{ic} = 0.4080\cdot 10^{-12}\left(\frac{\xi\alpha^a_{^3P}(\omega_m)I_r}{\Delta_{10}}\right)^2 w_{ic}, \qquad (9)$$

where $w_{ic}{=}1/\tau(\,^3P_1)$ is the field-free $^3P_1{\to}^1S_0$ intercombination transition rate, the data for which is presented in Table II (see e.g. [12, 13]).

As follows from the data of Table I, at the intensity $I_r{=}0.5\,MW/cm^2$, the absolute value of the magic-wave-induced amplitude (5) may amount to 10 *MHz* for atoms of Ca, Sr and Yb, that is equivalent to the amplitude induced by a magnetic field of 1 mT [6]. With account of the data for the spin-orbit splitting of the lowest (metastable) triplet state ^3P_J (see e.g. [14]) the admixture of the $^3P_1$ state in the $^3P_0$-state wave function at these conditions does not exceed 10^{-5}. Similar estimates indicate that the $^1P_1$ singlet state admixture in (7) at these conditions is yet 4 to 5 orders smaller.

TABLE I: Numerical values of the "magic" wavelength λ_{mag}, ${}^3P_1 - {}^3P_0$ splitting $\Delta_{10} = E_{{}^3P_1} - E_{{}^3P_0}$, anti-symmetric polarizability α^a_{3P} and the lattice-field-induced second-order Stark shift (lattice depth) $E_L^{(2)}$ for the ground-state and metastable alkaline-earth-like atoms in the optical lattice of the magic wavelength λ_{mag} and intensity I_L=10 kW/cm^2. The transition matrix element W_{10} is given for the mixing-wave intensity $I_r = 1\, MW/cm^2$.

Atom	λ_{mag} nm	Δ_{10} cm^{-1}	$\alpha^a_{3P}(\omega_m)$ $a.u.$	$E_L^{(2)}$ kHz	W_{10}/ξ MHz
Mg	432	20.06	538.5	−49.3	−10.3
Ca	680	52.16	−1054	−102	20.2
Sr	813.42[a]	186.83	−1044	−116	20.0
Yb	759.35[b]	703.57	−1084	−78.7	20.8
Zn	382	190.08	329.4	−21.3	−6.31
Cd	390	542.1	390.6	−25.8	−7.48

[a] the experimentally determined value [1, 2]
[b] the experimentally determined value [7]

TABLE II: Numerical values of the "clock" wavelength λ_c, coefficients $\kappa_p^{(1)}$ and $\kappa^{(2)}$ of linear in intensity of the probe field and quadratic in intensity of the circularly polarized lattice wave and/or mixing wave Stark shifts (12), the rate w_{ic} of spontaneous intercombination transition ${}^3P_1 \to {}^1S_0$ and the coefficient β for the Rabi frequency (10). The number in parentheses determines the power of ten.

Atom	λ_c nm	$\kappa^{(1)}(\omega_c)$ $\frac{mHz}{mW/cm^2}$	$\kappa^{(2)}(\omega_m)$ $\frac{Hz}{(MW/cm^2)^2}$	w_{ic} s^{-1}	$\lvert\beta\rvert$ $\frac{mHz}{MW/cm^2\sqrt{mW/cm^2}}$
Mg	458	4.27	-176	2.78(2)	32.7
Ca	660	−4.50	-255	2.94(3)	137.5
Sr	698	−44.2	-61.5	4.70(4)	176.9
Yb	578	24.5	-16.8	1.15(6)	180.6
Zn	309	0.816	-6.96	4.0(4)	15.2
Cd	332	23.0	-10.3	4.17(5)	22.6

However, the ${}^3P_1$-state admixture may be sufficient to enable the radiation transition between the ground and metastable states and to amplify the magnitude of the ${}^3P_0$ level width by 7 to 9 orders (in comparison with a two-photon E1-M1 or three-photon E1 spontaneous radiation decay width [10]), up to several mHz, making the clock transition ${}^1S_0 \to {}^3P_0$ well detectable, on the one hand, and on the other hand, retaining the ${}^3P_0$ level width in bosonic atoms essentially smaller than in fermionic.

Together with the radiative decay rate (9), the important characteristic of the magic-wave-induced ${}^1S_0 - {}^3P_0$ dipole transition, probed by the clock-frequency radiation, is the amplitude (Rabi frequency) of the clock transition which after integration in angular variables may be written as:

$$\Omega = \langle\, \psi | \hat{v}_p |\, {}^1S_0 \rangle = \beta I_r \sqrt{I_p}\; (i\, [\boldsymbol{e} \times \boldsymbol{e}^*] \cdot \boldsymbol{e}_p)\,, \qquad (10)$$

where $\hat{v}_p = \sqrt{I_p}(\boldsymbol{e}_p \cdot \boldsymbol{r})$ is the Hamiltonian of the dipole interaction between atom and probe field of intensity I_p and the unit polarization vector $\boldsymbol{e}_p$ which, evidently, should be parallel to the running wave vector $\boldsymbol{k} \propto i[\boldsymbol{e} \times \boldsymbol{e}^*]$, thus the maximal value of Ω will be for orthogonal propagation to the probe beam. So, in the case of a 1D optical lattice the Doppler-free interrogation is possible when the probe beam propagates along the lattice and is polarized along the mixing beam wave vector.

The coefficient β includes all radial integrals of the matrix element (10) which may be presented in the units of $mHz/(\sqrt{mW/cm^2} \cdot MW/cm^2)$, as follows:

$$\beta = 204.9\, \frac{\alpha^a_{3P}(\omega_m) \langle {}^1P_1^{(0)} | r |\, {}^1S_0 \rangle}{\Delta_{10}}\, b, \qquad (11)$$

with the antisymmetric polarizability and the radial part of the dipole transition matrix element in atomic units, the splitting Δ_{10} is in cm^{-1}. According to the calculated numerical values of β (see Table II), the Rabi frequency in Sr and Yb atoms (10) may achieve 0.3 Hz for the field (1) intensity I_r=0.5 MW/cm^2 and the probe field of I_p=10 mW/cm^2.

In the Stark-cancellation regime, when the second-order ac Stark shifts (4) of the clock levels are made equal to one another, the clock frequency may be distorted by the probe-field-induced quadratic ac Stark shift (linear in intensity $I_p \propto F_p^2$) and the fourth-order ac Stark shifts of the clock levels (quadratic in intensities $I_L \propto F_L^2$ and $I_r \propto F_r^2$, correspondingly), induced by the lattice field and the mixing wave, also including the bilinear in the intensities I_L and I_r fourth-order correction. This shift may be written as

$$\begin{aligned} \Delta\omega_c &= \kappa^{(1)}(\omega_c) I_p + \kappa^{(2)}(\boldsymbol{e}_L, \omega_m) I_L^2 + \kappa^{(2)}(\boldsymbol{e}, \omega_m) I_r^2 \\ &+ \kappa^{(2)}(\boldsymbol{e}_L, \boldsymbol{e}, \omega_m) I_L I_r, \end{aligned} \qquad (12)$$

where the constant $\kappa^{(1)}(\omega_c)$ is determined by the difference of the upper- and lower-level polarizabilities at the clock-transition frequency $\omega_c = 2\pi c/\lambda_c$. For $\kappa^{(1)}$ in the units of $mHz/(mW/cm^2)$ the relation is

$$\kappa^{(1)}(\omega_c) = -0.0469\, [\alpha_{{}^3P_0}(\omega_c) - \alpha_{{}^1S_0}(\omega_c)]\,, \qquad (13)$$

where polarizabilities $\alpha_{{}^3P_0}(\omega_c)$ and $\alpha_{{}^1S_0}(\omega_c)$ are in atomic units.

The coefficients $\kappa^{(2)}$ are determined by the difference of the clock-state hyperpolarizabilities at the "magic" frequency ω_m (similar to polarizabilities, the hyperpolarizabilities for states with the total momentum J=0 include only scalar parts, which, however, depend on the wave polarization vector $\boldsymbol{e}$ [9, 11]),

$$\begin{aligned} \kappa^{(2)}(\boldsymbol{e}, \omega_m) &= -8.359 \cdot 10^{-8} \\ &\times\; [\gamma_{{}^3P_0}(\boldsymbol{e}, \omega_m) - \gamma_{{}^1S_0}(\boldsymbol{e}, \omega_m)]\,, \end{aligned} \qquad (14)$$

where $\kappa^{(2)}$ is in the units of $Hz/(MW/cm^2)^2$, the hyperpolarizabilities $\gamma_{{}^1S_0}(\boldsymbol{e}, \omega_m)$ and $\gamma_{{}^3P_0}(\boldsymbol{e}, \omega_m)$ are in atomic

units. Although we assume the same "magic" frequency ω_m for the lattice and running waves, the hyperpolarizabilities for the linear polarization may differ essentially from those for the circular polarization [9, 11], i.e. $\kappa^{(2)}(\boldsymbol{e}_L, \omega_m) \neq \kappa^{(2)}(\boldsymbol{e}, \omega_m)$ for different polarization vectors $\boldsymbol{e}_L$ and $\boldsymbol{e}$. The clock-level hyperpolarizabilities determining the coefficient $\kappa^{(2)}(\boldsymbol{e}_L, \boldsymbol{e}, \omega_m)$ of the interference term, bilinear in the lattice-wave and running-wave intensities, depend on the relative orientation (and the type – linear or circular) of polarization vectors $\boldsymbol{e}_L$ and $\boldsymbol{e}$. That is why the fourth-order corrections from the both waves should be taken into account together with the mixed bilinear correction $\kappa^{(2)}(\boldsymbol{e}_L, \boldsymbol{e}, \omega_m) I_L I_r$.

The numerical estimates of $\kappa^{(1)}(\omega_c)$ and $\kappa^{(2)}(\boldsymbol{e}, \omega_m)$ for the circular polarization of the laser beam $\boldsymbol{e}$ are presented in table II. The hyperpolarizabilities for the metastable $^3P_0$ levels of the Mg, Zn and Cd atoms are complex values with imaginary parts (determining the two-photon ionization width) negligible in comparison with real parts. In estimating real parts of the hyperpolarizabilities in Mg, Zn and Cd, we took into account only the "resonant" terms, which may be determined by the antisymmetric and tensor polarizabilities of the levels [15].

The values of susceptibilities $\kappa^{(1)}$ and $\kappa^{(2)}$ of Table II are the useful data to control the higher-order corrections appearing when the probe-wave and running-wave intensities increase. However, the strong dependencies of $\kappa^{(2)}$ both on the polarization and on the frequency stimulate detailed investigations of the higher-order light shifts in the close vicinity of the magic-wave frequency (see e.g. [16, 17]). Given the values $\kappa^{(1)}$ and $\kappa^{(2)}$, all the combination of the probe-wave and higher-order shifts (12) becomes controllable and may in certain conditions be reduced to zero, using appropriate intensities I_L, I_r, I_p, and polarization $\boldsymbol{e}_L$ and $\boldsymbol{e}$ of the lattice and mixing waves.

In summary, we propose a new possibility to access the strongly forbidden single-photon 0-0 transition of a bosonic alkaline-earth-like atom supported by a magic-frequency wave with circular (elliptic) polarization. This approach may be considered as an alternative or an addition to the method of refs. [6], [7], where a magnetic field is used. It seems rather worthwhile for the lattice-based optical atomic clock with the magic-wave-supported strongly forbidden transition of even alkaline-earth-like isotopes. In the case of a 2D or 3D lattice, the running wave (1) may be replaced by one of the standing waves, which should be circular polarized and perpendicular to the probe laser beam. The circular polarization of the standing wave coincides with that of the incident wave whereas the amplitude near antinodes of the standing wave is twice as big as the incident-wave amplitude. In this case the space inhomogeneity of the mixing field should be taken into account, but if atoms occupy the lowest vibration states of the lattice and locate near antinodes then the field amplitude "seen" by an atom is double what it is in the incident wave, therefore the coefficients in the right-hand sides of equations (2), (5) and (10) can be multiplied by 4. It means that the Rabi frequencies (5) and (10) for the given laser input intensity may become 4 times greater for the standing wave in comparison with the running wave, in particular, in Sr and Yb atoms, W_{10}=40 MHz and Ω=1.2 Hz for I_r=0.5 MW/cm^2 and I_p=10 mW/cm^2.

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